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In re application of:

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Application No.: 10/668,964

Group Art Unit: 1711

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Examiner: Rabon Sergent

For: SOLVENT-FREE MOISTURE-CURABLE HOT MELT URETHANE RESIN
COMPOSITION

DECLARATION UNDER 37 CFR §1.132

COMMISSIONER FOR PATENTS

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Sir:

I, Yukihiko MINAMIDA, hereby declare and state that:

1. I am a citizen of Japan, residing at Dainippon Ink and Chemicals, Inc., 35-58, Sakashita 3-chome, Itabashi-ku, Tokyo, Japan.
2. I am one of the inventors of the subject application, and I am fully familiar with the subject matter thereof as well as the references relied upon by the Examiner in the prosecution of this application.
3. I obtained a Master's degree in Polymer Science and Engineering from the Kyoto Institute of Technology in March 1988.
4. I am currently employed by Dainippon Ink and Chemicals, Inc., and began

working for Dainippon Ink and Chemicals, Inc., on April 1, 1990, in research at the urethane synthesis laboratory. From April 2000, I have engaged in research with functional dispersion technology group.

5. I conducted the following experiments.

I. Object

With regard to the reference (United States Patent No. 4,574,793, United States Patent No. 5,616,625, United States Patent No. 5,550,191) which was cited in the Office Action (dated December 15, 2004) issued on the present application (U.S. Patent Application, No. 10/668,964), tests were conducted to provide supplemental data that the significant effect of the present invention can be obtained only when the polyester polyol having aromatic ring is used in combination with a morpholine ether-based crosslinking catalyst (B), sulfur atom-containing organic acid (C), and that such effect cannot be obtained using the polyol without aromatic ring as disclosed in the reference. It was shown that the polyol without aromatic ring in combination with a morpholine ether-based crosslinking catalyst (B), sulfur atom-containing organic acid (C) disclosed in the reference is not within the scope of claim of the present application.

II. Synthesis

In the following examples, parts are by mass unless otherwise specified.

[Comparative Example 1]

In four-necked flask, on which a stirrer, a condenser, and a dropping tube were mounted, 100 parts of methyl ethyl ketone, 17.8 parts of butyl methacrylate, 9.4 parts of methyl methacrylate, 0.068 parts of dodecyl mercaptan, and 1.7 parts of azo isobutyronitrile were heated to 80 °C under stirring. 94.1 parts of butyl methacrylate, 53.6 parts of methyl methacrylate, 0.612 parts of dodecyl mercaptan, and 100 parts of methyl ethyl ketone were added uniformly over a period of 1/2 hour while refluxing by heat of reaction and heating.

275.8 parts of polypropylene glycol having a number-average molecular weight

of 1000, 88.9 parts of polyester polyol having a number-average molecular weight of 3000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid, 29.3 parts of polyester polyol having a number-average molecular weight of 1000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid were heated to 100 °C under reduced pressure and then the de-solvent operation was performed until the methyl ethyl ketone was removed.

244 parts of 4, 4-diphenylmethane diisocyanate was added, and the reaction was held at 100 °C. After the 3 hours at 100 °C, 1.2 parts of 2, 2'-dimorpholine diethyl ether (trade name: "U-CAT660M", manufactured by SAN-APRO LTD) was added to obtain a solvent-free reactive hot melt urethane adhesive I having acryl-modified urethane prepolymer as main component. The solvent-free reactive hot melt urethane adhesive I has thermal stability melt viscosity of 9,000mPa·s/125 °C.

[Comparative Example 2]

In four-necked flask, on which a stirrer, a condenser, and a dropping tube were mounted, 100 parts of methyl ethyl ketone, 17.8 parts of butyl methacrylate, 9.4 parts of methyl methacrylate, 0.068 parts of dodecyl mercaptan, and 1.7 parts of azo isobutyronitrile were heated to 80 °C under stirring. 94.1 parts of butyl methacrylate, 53.6 parts of methyl methacrylate, 0.612 parts of dodecyl mercaptan, and 100 parts of methyl ethyl ketone were added uniformly over a period of 1/2 hour while refluxing by heat of reaction and heating.

275.8 parts of polypropylene glycol having a number-average molecular weight of 1000, 88.9 parts of polyester polyol having a number-average molecular weight of 3000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid, 29.3 parts of polyester polyol having a number-average molecular weight of 1000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid were heated to 100 °C under reduced pressure and then the de-solvent operation was performed until the methyl ethyl ketone was removed.

250 parts of 4, 4-diphenylmethane diisocyanate was added, and the reaction was held at 100 °C. After the 3 hours at 100 °C, 1.2 parts of U-CAT660M and 0.4 parts of methane sulfonate were added to obtain a solvent-free reactive hot melt

urethane adhesive II having acryl-modified urethane prepolymer as main component. The solvent-free reactive hot melt urethane adhesive II has thermal stability melt viscosity of 7,000mPa·s/125 °C.

[Comparative Example 3]

In four-necked flask, on which a stirrer, a condenser, and a dropping tube were mounted, 100 parts of methyl ethyl ketone, 17.8 parts of butyl methacrylate, 9.4 parts of methyl methacrylate, 0.068 parts of dodecyl mercaptan, and 1.7 parts of azo isobutyro nitrile were heated to 80 °C under stirring. 94.1 parts of butyl methacrylate, 53.6 parts of methyl methacrylate, 0.612 parts of dodecyl mercaptan, and 100 parts of methyl ethyl ketone were added uniformly over a period of 1/2 hour while refluxing by heat of reaction and heating.

275.8 parts of polypropylene glycol having a number-average molecular weight of 1000, 88.9 parts of polyester polyol having a number-average molecular weight of 3000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid, 29.3 parts of polyester polyol having a number-average molecular weight of 1000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid, and 380 parts of an aromatic polyester polyol obtained by using an isophthalic acid and a terephthalic acid (trade name: "Dynacoll 7130", manufactured by SAN-APRO LTD) were heated to 100 °C under reduced pressure and then the de-solvent operation was performed until the methyl ethyl ketone was removed.

300 parts of 4, 4-diphenylmethane diisocyanate was added, and the reaction was held at 100 °C. After the 3 hours at 100 °C, 1.9 parts of U-CAT660M was added to obtain a solvent-free reactive hot melt urethane adhesive III having acryl-modified urethane prepolymer as main component. The solvent-free reactive hot melt urethane adhesive III has thermal stability melt viscosity of 14,000mPa·s/125 °C.

[Example]

In four-necked flask, on which a stirrer, a condenser, and a dropping tube were mounted, 100 parts of methyl ethyl ketone, 17.8 parts of butyl methacrylate, 9.4 parts of methyl methacrylate, 0.068 parts of dodecyl mercaptan, and 1.7 parts of azo isobutyro

nitrile were heated to 80 °C under stirring.

94.1 parts of butyl methacrylate, 53.6 parts of methyl methacrylate, 0.612 parts of dodecyl mercaptan, and 100 parts of methyl ethyl ketone were added uniformly over a period of 1/2 hour while refluxing by heat of reaction and heating. 275.8 parts of polypropylene glycol having a number-average molecular weight of 1000, 88.9 parts of polyester polyol having a number-average molecular weight of 3000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid, 29.3 parts of polyester polyol having a number-average molecular weight of 1000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid, and 380 parts of Dynacoll 7130 were heated to 100 °C under reduced pressure and then the de-solvent operation was performed until the methyl ethyl ketone was removed.

300 parts of 4, 4-diphenylmethane diisocyanate was added, and the reaction was held at 100 °C. After the 3 hours at 100 °C, 1.9 parts of U-CAT660M and 0.5 parts of methane sulfonate were added to obtain a solvent-free reactive hot melt urethane adhesive IV having acryl-modified urethane prepolymer as main component. The solvent-free reactive hot melt urethane adhesive IV has thermal stability melt viscosity of 12,000mPa·s/125 °C.

[Comparative Example 4]

In a 1 liter four-necked flask, 700 parts of long chain aliphatic polyester polyol having a number-average molecular weight of 3500 obtained by reacting 1,2-dodecane dicarboxylic acid with 1,6-hexanediol were heated to 100 °C under reduced pressure and then dehydrated until the water content was reduced to 0.05%.

After cooling to 70 °C, 120 parts of diphenylmethane diisocyanate was added and the mixture was heated to 100 °C and was reacted for 3 hours while being maintained until the isocyanate group content became constant to obtain an aliphatic polyester-urethane prepolymer.

After the completion of the reaction, 1.6 parts of U-CAT 660M and 0.4 parts of methane sulfonic acid were added and stirred uniformly to obtain a solvent-free moisture-curable hot melt urethane resin composition V having an aliphatic polyester-urethane prepolymer as main component. Properties of the resulting

solvent-free moisture-curable hot melt urethane resin composition V has thermal stability melt viscosity of 15,000mPa·s/125 °C.

[Comparative Example 5]

In a 1 liter four-necked flask, 700 parts of polypropylene glycol having a number-average molecular weight of 3000 was heated to 70 °C under reduced pressure and then dehydrated until the water content was reduced to 0.05%.

100 parts of carbodiimide-modified diphenylmethane diisocyanate was added and the mixture was heated to 100 °C and was reacted for 3 hours while being maintained until the isocyanate group content became constant to obtain an aliphatic polyether-urethane prepolymer.

After the completion of the reaction, 1.6 parts of U-CAT 660M and 0.4 parts of methane sulfonic acid were added and stirred uniformly to obtain a solvent-free moisture-curable hot melt urethane resin composition VI having an aliphatic polyether-urethane prepolymer as main component. Properties of the resulting solvent-free moisture-curable hot melt urethane resin composition VI has thermal stability melt viscosity of 1,000mPa·s/125 °C.

III. Tests

[Method for evaluation of thermal stability]

A solvent-free moisture-curable hot melt urethane resin composition was charged in a metal can having an inner volume of 200 ml and allowed to stand in an oven at 120 °C in a sealed state for 18 hours and 36 hours. Then, it was visually observed whether gelation occurred or not. In the case in which no gelation occurred after standing, the melt viscosity (mPa·s) at 125 °C was measured.

[Method for evaluation of creep resistance at high temperature]

The creep resistance at high temperature was evaluated by the following procedure in place of the evaluation of the moisture-crosslinking reactivity at room temperature.

Using a specimen made by laminating an olefin decorative sheet and a middle

density fiber (MDF) board via a solvent-free moisture-curable hot melt urethane resin composition, the creep resistance at high temperature was evaluated in the following manner.

A solvent-free moisture-curable hot melt urethane resin composition melted heat-melted on a hot plate having a surface temperature of 80°C was applied at a thickness of 40 μm on a 50 μm thick olefin decorative sheet and a MDF board was laminated on the coated surface and, after roll pressing at a linear pressure of 10 kg/cm, the resulting laminate was cut into a width of 25 mm to prepare for a bonded specimen.

A heat-resistant creep test was carried out by allowing specimens, which were allowed to stand for 1, 2, 3, or 4 hours immediately after production at room temperature (23 °C, relative humidity: 65%), to stand in an oven at 60°C for 1 hour while perpendicularly suspending a load (500 g/25 mm) at one end of the olefin decorative sheet in a 90°C angle peeling direction. Then, a creep distance (mm) from the initial suspending location was measured over time. Specimens which had the creep distance of no more than 5 mm were rated "good".

Since the initial creep properties at 60 °C are improved due to proceeding of the crosslinking reaction at room temperature (23 °C, relative humidity: 65%), the initial creep properties were evaluated in place of the moisture-crosslinking reactivity at room temperature.

IV. Test Results

The test results are shown in the following Table.1.

Table.1

	Comparative Example 1	Comparative Example 2	Comparative Example 3	Example 1	Comparative Example 4	Comparative Example 5
Thermal stability						
Melt viscosity (mPa·s, 125°C)						
initial	9,000	7,000	14,000	12,000	15,000	1,000
after 18 hours	9,400	8,100	22,000	14,000	15,000	1,000
after 36 hours	10,300	9,600	Gelled	22,000	19,000	2,000
Creep resistance at 60°C						
Immediately after production of specimen	Fell off	Fell off	Fell off	Fell off	Fell off	Fell off
1 hour after standing at room temperature	Fell off	Fell off	Fell off	Fell off	Fell off	Fell off
2 hours after standing at room temperature	Fell off	Fell off	Fell off	Fell off	Fell off	Fell off
3 hours after standing at room temperature	Fell off	Fell off	26mm	35mm	Fell off	Fell off
4 hours after standing at room temperature	Fell off	Fell off	Good	Good	Fell off	Fell off
6 hours after standing at room temperature	Fell off	Fell off	Good	Good	Fell off	Fell off
8 hours after standing at room temperature	Fell off	Fell off	Good	Good	Fell off	Fell off
12 hours after standing at room temperature	Good	Fell off	Good	Good	Fell off	Fell off

*Regarding property of creep resistance at high temperature of Comparative Example 4 and 5, low adhesive strength cause "Fell off".

V. Conclusion

As indicated above, the solvent-free moisture-curable hot melt urethane resin composition, in which the polyester polyol having aromatic ring is used in combination with a morpholine ether-based crosslinking catalyst (B), sulfur atom-containing organic acid (C), has significant effects as claimed in claim 1 of the present application.

We believe, in addition to the significant structural difference between the present invention and the reference, that it is demonstrated the urethane prepolymer disclosed in the reference is not within the scope of claim 1 of the present application.

6. I understand fully the content of this declaration.

7. The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

8. Further declarant saith not.

Yukihiko Minamida

Yukihiko MINAMIDA

2005. 4. 12

Date

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